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AIR FORCE OFFICE OF SCIENTIFIC RESEARCH

Final Progress Report

FOR

01 March 1998 through 28 February 2001

CONTRACT: AFOSR

F49620-98-1-0330 (AASERT 98)

TITLE OF CONTRACT: Infrared Material Development based on III-V

Antimonides

NAMES OF PRINCIPAL INVESTIGATORS: Wen I. Wang, Professor

NAME OF ORGANIZATION: Electrical Engineering Department,

Columbia University

ADDRESS OF ORGANIZATION: Electrical Engineering Department, Columbia University, 500 West 120th Street, New York, N.Y. 10027

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- 1. Objective: Develop infrared materials based on III-V antimonides grown by molecular beam epitaxy (MBE).
- 2. Status of effort: Near Infrared optical Investigation of InGaAsN with Sb has been carried out. Detailed absorption and photoreflectance have been measured. X-ray diffraction of the materials has been performed to ascertain the crystallinity of the material. Sb was used as a surfactant for the growth of GaN. Photoluminescence and x-ray diffraction both showed improvement compared to samples grown without Sb.

3. Accomplishments/New Findings:

Two papers were presented at the SPIE Conference, Photodetectors: Materials and Devices, 2001, and the North American Conference on Molecular Beam Epitaxy, 2000. The papers were published in the Proceedings of the SPIE, vol. 4288 and J. Vac. Sci. and Tech. 2001.

4. Personnel Supported:

Graduate Research Student working under this project: Brandon Turk (US Citizen)

5. Publications:

- C.W. Pei, B. Turk, J. Heroux, and W.I. Wang, "GaN grown by molecular beam epitaxy with antimony as surfactant", J. Vac. Sci. and Tech. B19, 1426 (2001).
- J. Heroux, X. Yang, B. Turk, and W.I. Wang, "Optical investigation of InGaAsN structures for photodetector applications", Proceedings of SPIE, vol. 4288 (2001).
- 6. Interactions/Transitions: Presentations at conferences
- C.W. Pei, B. Turk, J. Heroux, and W.I. Wang, "GaN grown by molecular beam epitaxy with antimony as surfactant", presented at the North American Conference on Molecular beam epitaxy, Oct 16, 2000, Arizona State University, AZ.
- J. Heroux, X. Yang, B. Turk, and W.I. Wang, "Optical investigation of InGaAsN structures for photodetector applications", presented at the SPIE Conference, Jan. 22, 2001, San Jose, CA.
- 7. New discoveries, inventions, or patent disclosures: None

Optical Investigation of InGaAsN Structures for Photodetector Applications

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ABSTRACT

The optical properties of $\ln_{1.5}Ga_{.85}As_{1.x}N_x$ structures for the fabrication of photodetectors are investigated. An expression for the bulk bandgap as a function of the nitrogen fraction is obtained from x-ray diffraction, photoreflectance and photoluminescence measurements. Optical absorption of undoped MQW structures show that the cutoff wavelength is extended due to the presence of nitrogen. A functioning heterojunction phototransistor was fabricated. Photocurrent spectra show that a responsivity higher than 1.5 A/W is obtained with a cutoff wavelength of 1.16 μ m. I-V measurements under different light levels show that a peak gain of 5 is obtained with a collector current of 260 μ A and a dark current lower than 2 μ A with a 10V bias.

Keywords: Phototransistor. InGaAsN. multi-quantum wells, absorption, photoreflectance, photoluminescence, photocurrent, x-ray diffraction.

1. INTRODUCTION

Recently a new quaternary material, InGaAsN, has attracted a lot of attention due to its potential for the fabrication of high performance GaAs-based optoelectronic devices operating in the near infrared. Adding a small fraction of nitrogen to the In_xGa_{1.x}As ternary compound leads to a decrease in both the bandgap and the lattice constant, hence increasing flexibility in device design. However, the growth of high quality, high nitrogen content InGaAsN expitaxial layers has been a challenge, and for nearly all devices demonstrated so far operating near the technologically important 1.3 µm wavelength, a compressively strained InGaAsN active region was used.

For photodetector applications, the new quaternary material offers the possibility of growing resonant cavity enhanced (RCE) devices operating in the 1.3-1.55 µm wavelength range on high reflectance, lattice-matched GaAs/AlAs distributed Bragg reflectors [1, 2]. However, if compressive strain is advantageous in the case of InGaAsN single quantum well lasers to provide a better carrier confinement, it is detrimental in photodetectors where a thicker active region is needed to obtain the desired absorption. It has been shown that the dark current in p-i-n structures grown beyond critical thickness is highly sensitive to the dislocation density [3].

A promising candidate for the fabrication of high responsivity, low dark current photodetectors with a strained active region is the heterojunction phototransistor (HPT) with a multi-quantum well (MQW) absorbing region[4]. Ghisoni et al.[5.6] have shown that a slight relaxation in this type of structure can be tolerated and does not result in catastrophic device failure. Therefore, a higher lattice mismatch between layers can be tolerated and the fabrication of devices with a longer operating wavelength on GaAs substrates is possible.

The purpose of this paper is to perform a detailed optical characterization of compressively strained InGaAsN layers for photodetector applications. The operation of an InGaAsN HPT with a cutoff wavelength of 1.16 µm is demonstrated.

2. EXPERIMENTS

Samples were grown by solid-source Molecular Beam Epitaxy using a radio-frequency (rf) N radical beam source (SVT Associates) [7]. Three different types of InGaAsN structures were grown. In all cases, the nominal indium concentration was chosen to be 15%. First, single layers with nitrogen concentrations varying from 0 to 2% were grown to estimate the bandgap reduction as a function of nitrogen. The nominal thickness of these single layer samples was 200 Å to avoid relaxation of the epitaxial layer. Undoped In 15Ga_{.85}As_{1-x}N_x/GaAs multi-quantum well structure (10 periods, 80 Å well/200 Å barrier nominal thickness) were also grown to estimate the average strain and absorption coefficient as a function of nitrogen concentration. Finally, an npin heterojunction phototransistor structure with a wide-band-gap emitter and an InGaAsN/GaAs MQW absorbing intrinsic collector region was grown (figure 1). The MQW collector region had 20 periods to ensure a high absorption. Antimony was used as a surfactant to improve the material quality of the MQW[8]. Mesas were etched using conventional lithography techniques and a Au-Ge-Ni alloy for ohmic contacts.

				Dopin	g (cm ⁻³)	Thick (A
		Cap	GaAs	ή+	2E+18	1000
		Emitter	Al.3Ga.7As	n	1.40E+18	1000
		Emitter	Al.3Ga.7As	n	5E+17	2000
20X [Base	GaAs	P	5E+17	1000
		Spacer	GaA5	i		2575
	Г	Barrier	GaAs	i		150
		Well	GalnNAs:Sb	i		85
	Ĺ	Barrier	GaAs	i		150
		Spacer	GaAs	<u>i</u>		2575
		Subcollector	GaAs	n+	2E+18	10000
		Substrate	GaAs	n+		

Figure 1 InGaAsN npin heterojunction phototransistor structure.

Structural analysis was done by x-ray diffraction in the 400, 511+ and 511- orientations using a five crystals diffractometer. A Bomem DA8 Fourier Transform Spectrometer equipped with a Quartz halogen light source and InGaAs and Ge photodetectors was used for room temperature optical characterization by photoluminescence, absorption and photocurrent. Absolute responsivity values for the phototransistor structure were obtained by comparing the response of 300 μ m diameter mesas to that of a calibrated Ge photodiode with the same active area. Photoreflectance was done with a 0.5 meter focal length grating spectrometer, a quartz halogen light source, an InGaAs photodiode and a 670 nm laser diode pulsed at 1 kHz with a light power of 1 mW. I-V characterization of the phototransistor structure under varying light power was done using an HP 4145B semiconductor parameter analyzer and a high power laser diode emitting at 980 nm focused on the 50X100 μ m² optical window of 50 X 150 μ m² mesas. The wavelength of the laser diode was chosen to be above the GaAs substrate bandgap and below the InGaAsN MQW absorption edge to ensure absorption in the collector region of the device only.

3. RESULTS AND DISCUSSION

3.1 Bulk samples

An accurate expression of the bulk bandgap as a function of indium and nitrogen concentrations is needed in order to properly design MQW structures. Figure 2 shows photoreflectance and photoluminescence spectra of 200 Å bulk In₁₆Ga₈₆As_{1.3}N, samples with increasing nitrogen concentration from top to bottom. A third derivative functional form curve fit from which the fundamental transition was obtained is superposed on each experimental photoreflectance curve. For the last two samples, only a PL signal could be detected. The indium and nitrogen concentrations were estimated by performing 400 and 511 x-ray diffraction analysis. In each case, the sample was

rotated 180° and the layer peak position was taken to be the average of the two orientations to take into account possible tilt of the epitaxial layer with respect to the substrate [9].

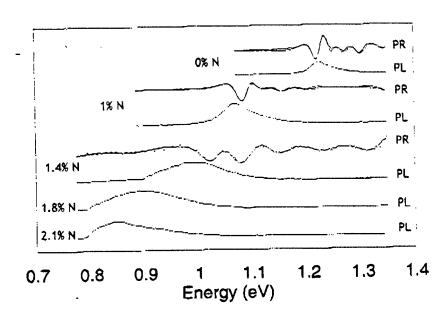


Figure 2: Room temperature photoreflectance (first three samples only) and photolumnescence spectra of 200 Å bulk $\ln_{16}Ga_{34}As_{1-3}N_x$ layers. A third derivative functional form curve fitting is shown on top of the experimental photoreflectance curves.

Figure 3 summarizes the experimental results. A decrease of the bandgap is clearly observed as the nitrogen composition is increased. The values obtained by photoreflectance and photoluminescence for the first three samples are within reasonable agreement, indicating that photoluminescence is an accurate way to determine the bandgap in these structures [10,11].

The expression we used to evaluate the bulk bandgap is similar to the one used by Tanaka et al.[10]:

$$Eg(In_{x}Ga_{1-x}As_{1-y}N_{y}) = y \cdot E_{g}(In_{x}Ga_{1-x}N) + (1-y) \cdot E_{g}(In_{x}Ga_{1-x}As) - b_{x}y \cdot (1-y) + \delta E(strain)$$
 (1)

The bandgap of $In_xGa_{1.x}N$ was calculated assuming a value of 3.50 eV for the bandgap of zinc-blende GaN [12] and 1.8 eV for InN with a bowing parameter of 1.4 eV [13]. The value for the unstrained $In_xGa_{1.x}As$ bandgap was obtained using the expression given by Nahory et al.[14]. A term $\delta E(strain)$ was included to take the compressive strain into account assuming Vegard's law. Values for the deformation potentials and elastic constants were those given by Arent et al [15] for $In_xGa_{1.x}As$ and it was assumed that they were not affected by the presence of a small nitrogen fraction.

As shown in figure 3, a reasonably accurate fit of our data is obtained assuming $b_x=18$ eV, a value in the range of those reported for $GaAs_{1,x}N_y$.[16]. This suggests that the bowing parameter of $In_xGa_{1,x}As_{1,x}N_y$ may not be significantly altered by the presence of indium atoms if the bulk bandgap increase due to compressive strain is taken into account.

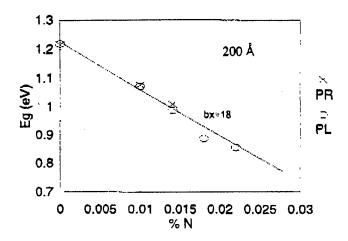


Figure 3: Fundamental transition as a function of the nitrogen concentration in bulk $In_{.16}Ga_{.84}As_{1..x}N_x$ samples acquired by photoluminescence and photoreflectance measurements. The line shows a curve fit obtained using equation (1) with $b_x=18$ eV.

3.2 Multi-quantum well structures

Figure 4 shows x-ray diffraction spectra of MQW samples with varying nitrogen concentrations. The average strain, period, well width and atomic concentrations were estimated using a dynamical theory simulation [17] assuming that the indium content did not vary among samples. The well width and period were found to be within 2 monolayers of their nominal values for all samples. The period being nearly constant, a decrease in average strain as the nitrogen concentration is increased can be clearly seen in figure 4.

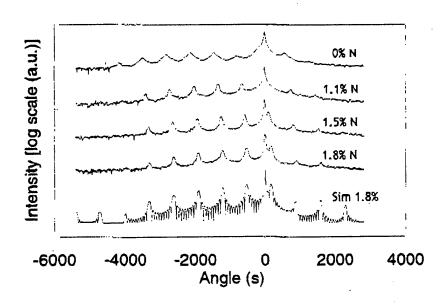


Figure 4: X-ray diffraction spectra (top 4 curves) and simulation (bottom curve) of $In_{15}Ga_{89}As_{1-x}N_x/GaAs$ multi-quantum wells with varying nitrogen content.

Figure 5 shows the absorption of the multi-quantum well samples. A shift to a longer wavelength is clearly observed as the nitrogen concentration is increased, showing the potential of this material for the fabrication of near infrared photodetectors. A normalized absorption coefficient αd near 0.1, a typical lower boundary value for the fabrication of high quantum efficiency RCE devices [18], was obtained for all 10 periods samples above the fundamental transition.

The inset shows the variation of the fundamental transition energy as a function of the nitrogen percentage. A calculation of the quantum confined energy levels assuming a bulk bandgap given by equation (1) with $b_x=16$ eV and a strained band offset of 0.7 shows that the fundamental transition for the two higher concentration cases is higher than expected. The reason for this discrepancy is unclear at present and could be due to uncertainties in the determination of the atomic concentrations or well width.

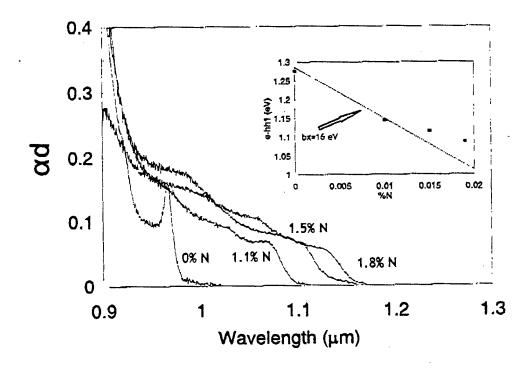


Figure 5: Normalized absorption spectra of 10 period multi-quantum well samples with varying nitrogen content. Inset shows fundamental transition energies.

3.3 Heterojunction phototransistor device

A photocurrent spectra of the phototransistor device is shown in figure 6 at different biases. A responsivity higher than 1.5 A/W was measured with a wavelength cutoff of 1.16 µm with a 10V bias, clearly indicating that the device exhibits gain. However, as shown in the figure, the responsivity decreased rapidly for lower biases. Since the spectra was acquired with an FTIR, a fraction of the incident light was not modulated so that the sample was under white light illumination during the measurement. At a lower bias, weak exciton features can be observed.

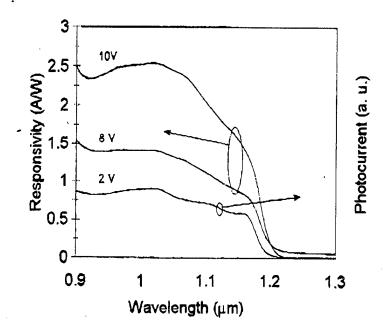


Figure 6: Photocurrent spectra of an InGaAsN/GaAs heterojunction phototransistor at 10V, 8V and 2V collector-emitter biases.

Figure 7 shows the collector current, I_C , as a function of the applied collector-emitter bias V_{CE} for different incident light powers. Under all incident light powers, a clear saturation level is reached, an essential feature of a good phototransistor. Moreover, the dark current was less than 2 nA, nearly two orders of magnitude lower than the photocurrent under a 1 μ W incident light power. It did not increase significantly with the bias, showing the good quality of the sample. These features compare advantageously with the results reported by Ghisoni et al. [4,5], who have shown that partially relaxed phototransistors tend not to reach a clear saturation level and have a non-negligible dark current at as the collector-emitter bias voltage is increased. However, our device exhibited a high turn-on voltage, which could be due to an incomplete carrier collection efficiency caused by the high conduction band offset of the InGaAsN/GaAs MQWs[4].

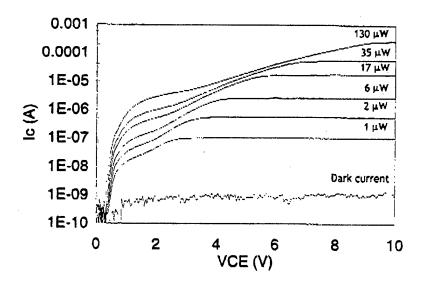


Figure 7: Collector current I_c as a function of the collector-emitter voltage V_{CE} for different incident optical powers.

The gain of the phototransistor, given by

$$G = \frac{hc}{q\lambda} \frac{I_c}{P_{inc}} \tag{2}$$

is shown in figure 8 as a function of the collector current on a log-log scale. A peak gain of 5 is obtained with a collector current of 260 μ A. A nearly linear relationship is obtained for a lower collector current I_c , from which the ideality factor of the emitter-base junction n can be estimated using the relation

$$G \propto I_c^{(1-1/n)} \tag{3}$$

A value of n=1.5 is obtained, showing the good quality of the emitter-base junction. However, figure 8 shows that the gain is slightly sub-linear and tends to saturate at a high collector current I_c , which may indicate that the generation-recombination component at the emitter is not the dominant defect current[19]. This estimation of the ideality factor n also assumes that the base transport factor η_b is near unity, which may not be an accurate assumption if the minority carrier diffusion length in the base is small.

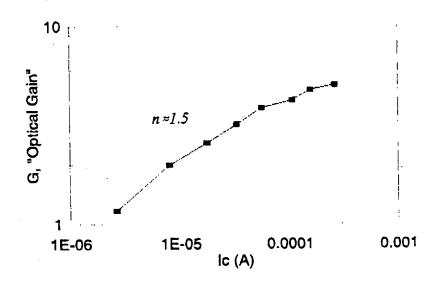


Figure 8: Heterojunction phototransistor optical gain as a function of the collector current L.

4. Summary

In summary, we have investigated the optical properties of $\ln_{15}Ga_{85}As_{1-7}N_x$ structures for the fabrication of photodetectors. An expression for the bulk bandgap as a function of the nitrogen fraction was obtained from photoreflectance and photoluminescence measurements. Optical absorption of undoped MQW structures show that the cutoff wavelength is extended due to the presence of nitrogen. A normalized absorption coefficient αd close to 0.1 is obtained above the fundamental transition with a 10 period structure. A functioning heterojunction phototransistor with a cutoff wavelength of 1.16 μm , a peak gain of 5 and a dark current lower than 2 nA with a 10 V collector-emitter bias has been demonstrated.

5. Acknowledgments

B. Turk was supported by AFOSR AASERT (Dr. Dan Johnstone). W. I. Wang acknowledges the support of a Guggenheim Fellowship.

6. References

- 1. J.B. Héroux, X. Yang and W.I. Wang, Appl. Phys. Lett. 75 (18) 2716 (1999)
- 2. G.S. Kinsey, D.W. Gotthold, A.L. Holmes, Jr., and J.C. Campbell, Appl. Phys. Lett. 77 (10) 1543 (2000).
- 3. J.P.R. David, Y.H. Chen, R. Grey, G. Hill. P.N. Robson and P. Kightley, Appl. Phys. Lett. 67 (7), 906 (1995).
- 4. R. P. Bryan, G.R. Olbright, W.S. Fu. T.M. Brennan and J.Y. Tsao, Appl. Phys. Lett. 59 (19), 1600 (1991).
- 5. M. Ghisoni, O. Sjölund, A. Larsson, J. Thordson, T. Andersson, S.M. Wang, and L. Hart, IEEE J. Sel. Top. Quant, Elect. 3 (3), 768 (1997).
- 6. M. Ghisoni, O. Sjölund, and A. Larsson, Appl. Phys. Lett. 69 (12), 1773 (1996).
- 7.X. Yang, J.B. Héroux, M.J. Jurkovic and W.I. Wang, J. Vac. Sci. Technol. B 17, 1144 (1999).
- 8. X. Yang, M.J. Jurkovic, J.B. Héroux, and W.I. Wang, Appl. Phys. Lett. 75 (2), 178 (1999).
- 9. A.T. Macrander, G.P. Schwarz, and G.J. Gualtieri, J. Appl. Phys. 64 (12), 6733 (1999).
- 10. S. Tanaka, M. Takahashi, A. Moto, T. Tanabe, S. Takagishi, K. Karatani, T. Nakahishi, and M. Nakayama, Inst. Phys. Conf. Ser. 162, ch. 11, 633 (1999).
- 11. M. Hetterich, M.D. Dawson, A. Yu. Egorov, D. Bernklau, and H. Riechert, Appl. Phys. Lett. 76 (8), 1030 (2000).
- 12. A.F. Wright and J.S. Nelson, Appl. Phys. Lett. 66 (22), 3051 (1995).
- 13. R. Goldhahn, J. Scheiner, S. Shokhovets, T. Frey. U. Kohler, D.J. As, and K. Lischka, Appl. Phys. Lett. 76 (3), 291 (2000).
- 14. R.E. Nahory, M.A. Pollack, W.D. Johnston Jr., and R.L. Barns, Appl. Phys. Lett. 53, 2510 (1988).
- 15. D.J. Arent. K. Deneffe. C. Van Hoof, J. De Boeck, and G. Borghs, J. Appl. Phys. 66 (4), 1739 (1999).
- 16. S. Wei and A. Zunger, Phys. Rev. Lett. 76 (4), 664 (1996).
- 17. C.R. Wie, Mat. Sci. Eng. R13 (1), 1 (1994).
- 18. M.S. Unlü and S. Strite, J. Appl. Phys. 78 (2), 607 (1995).
- 19. J.C. Campbell, Semiconductors and Semimetals (Academic, Orlando, 1985), Vol. 22, part D.

GaN grown by molecular beam epitaxy with antimony as surfactant

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In this work, we investigated the effect of Sb on the molecular beam epitaxy MBE growth of GaN and its optical properties. One monolayer Sb predeposition before GaN growth and different Sb beam equivalent pressures applied throughout the GaN growth were used to study the effect of Sb on GaN growth by ammonia gas-source MBE. The presence of Sb remarkably enhanced two-dimensional growth as evidenced by in situ reflected high energy electron diffraction RHEED. RHEED patterns became streaky much more rapidly when GaN was grown in the presence of Sb than that without Sb, indicating that Sb can act as an effective surfactant to smooth the growth front of GaN and enhance the layer-by-layer growth mode for the MBE growth of GaN. The full width at half maximum of the 0004 x-ray diffraction rocking curve measured from the GaN epilayer grown with one monolayer Sb predeposited as surfactant was as narrow as 70 arcsec. In the photoluminescence measurement, besides the characteristic near band edge excitonic emissions, new transitions related to Sb-isovalent traps were observed from GaN samples grown with Sb, whose zero phonon line was located at 3.27 eV with phonon replicas on the lower energy side. Intensities of the emissions related to Sb-isovalent traps increased with Sb partial pressures applied during GaN growth. © 2001 American Vacuum Society. DOI: 10.1116/1.1374627

I. INTRODUCTION

GaN has attracted great attention due to its potential applications in high-power, high-temperature microelectronic devices and optoelectronic devices operating in the blue to ultraviolet spectral region. 1,2 Although high quality GaN has been obtained by metalorganic chemical vapor deposition MOCVD, molecular beam epitaxy MBE is a viable alternate growth method with the advantage of accurate in situ thickness control at the monolayer scale monitored by reflection high energy electron diffraction RHEED. 3 It is a challenge to grow device-quality GaN epilayers by MBE because of the large lattice mismatch between GaN and the sapphire substrate. The lattice mismatch introduces strain into the film and leads to the Volmer-Weber or Stransky-Kastanow growth mode, in which three-dimensional 3D islands are formed to relax the strain and a high density of defects is introduced. In order to achieve high quality GaN epilayers, it is desirable to have a layer-by-layer growth mode by twodimensional step propagation. A two-step growth process, 4-9 which consisted of a thin AlN or GaN buffer layer of 20-30 nm grown at low temperature (500-600 °C), has been found to improve the quality of GaN epilayers. In addition, surfactants have been used to improve crystal growth. Previously, we used Sb as a surfactant to improve the epitaxial growth of InGaAsN. 10,11 For GaN growth by MBE, indium has been employed as a surfactant to enhance layer-by-layer growth. 12 Surfactant atoms can saturate surface dangling bonds and chemically passivate the surface. As a result, the energy of the system is lower when the surfactant atoms are at the growth front rather than being buried. In this work, we used Sb as surfactant for the growth of GaN by MBE, and investigated the effect of Sb on GaN growth using in situ RHEED.

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The influence of Sb on the optical properties of GaN was studied by low-temperature photoluminescence PL measurements for a series of samples grown in the presence of different Sb partial pressures.

II. EXPERIMENTS

The samples were grown by gas-source molecular beam epitaxy GSMBE . High purity ammonia gas was used as the nitrogen source. Ga and Al were supplied from conventional Knudsen effusion cells. In this study, all GaN layers were grown on basal plane sapphires, which are readily available. Before GaN growth, the sapphire substrate was heated to 900 °C for outgassing, followed by a 20 nm AlN buffer layer deposited at 810 °C. Then a GaN epilayer was grown with one monolayer Sb predeposited before its growth. This film was compared with the GaN epilayers grown with Sb beam equivalent pressures of 0.8×10^{-7} , 1.6×10^{-7} , 2.1×10^{-7} , and 3.3×10^{-7} Torr, respectively, applied throughout the entire growth of GaN. In GaN growth, a NH3 flow rate of 60 sccm and a Ga beam equivalent pressure of 1.1×10^{-6} Torr were used. The thickness of all GaN epilayers was 2.5 μ m. RHEED was employed for in situ monitoring. To investigate the effect of Sb on the optical properties of GaN, lowtemperature PL measurements were carried out with the 325 nm line of a He-Cd laser as an excitation source. To facilitate comparison, all measurements were under the same pump power.

III. RESULTS AND DISCUSSIONS

The RHEED pattern was spotty for GaN grown on an AlN nucleation layer without Sb. But when GaN was grown in the presence of Sb, the RHEED pattern changed from spotty to streaky dramatically. Figure 1 shows the RHEED patterns of GaN epilayers grown with and without Sb to the

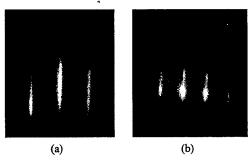


Fig. 1. Comparison of GaN RHEED patterns: a 20 nm GaN overgrowth with Sb as surfactant and b 20 nm GaN overgrowth without Sb.

same thickness of 20 nm. The RHEED pattern is streaky for GaN grown with Sb, in sharp contrast to the spotty pattern of GaN grown without Sb, shown in Fig. 1 b. The (2×1) RHEED pattern observed along the 1120 azimuth during GaN growth indicated that Sb greatly facilitated the twodimensional growth mode. Figure 2 shows the 0004 x-ray diffraction XRD rocking curves of GaN epilayers. For the GaN epilayer grown with one monolayer Sb predeposited before GaN growth as surfactant, the full width at half maximum FWHM was 70 arcsec and was much narrower than that of GaN grown without Sb, indicating superior crystallinity. Our results clearly suggest that Sb can act as an effective surfactant for GaN growth. For the samples which were grown with a Sb beam flux applied throughout the entire GaN growth, the FWHM is broader than that of GaN grown with one monolayer Sb predeposited as surfactant. This was due to the deterioration of crystal quality upon excess Sb incorporation into GaN when a Sb beam flux was applied throughout the entire growth of GaN.

PL measurements were carried out at 10 K for a series of GaN samples grown in the presence of different Sb beam equivalent pressures of 0.8×10^{-7} , 1.6×10^{-7} , 2.1×10^{-7} , and 3.3×10^{-7} Torr, respectively, applied throughout the entire growth of GaN. Figure 3 shows the PL spectra of GaN epilayers grown with and without Sb. Yellow band emission related to deep-level impurities and defects was absent. Excitonic emission EE at 3.47 eV and its weak phonon replica at 3.37 eV can be clearly identified. A significant increase of

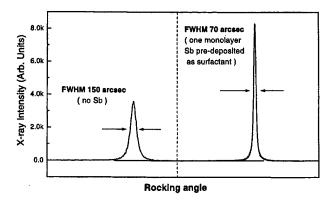


Fig. 2. 0004 XRD rocking curves of GaN films grown with one monolayer predeposited Sb as surfactant and without Sb.

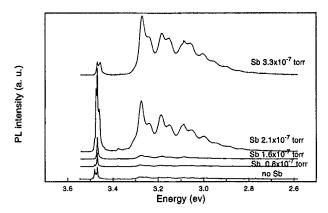


Fig. 3. Photoluminescence spectra of GaN epilayers grown in the presence of different Sb beam fluxes by GSMBE.

band edge emission intensity was exhibited with Sb beam flux up to 2.1×10^{-7} Torr. An intense band edge luminescence was also observed from a GaN epilayer grown with indium as surfactant by another group. The near band edge emission intensity decreased when the Sb beam equivalent pressure was higher than 3.3×10^{-7} Torr. This could be due to the introduction of defects when excess Sb atoms were incorporated in the GaN epilayers.

New transitions were observed in photoluminescence from GaN samples grown with Sb, whose zero phonon line ZPL was located at 3.27 eV with phonon replicas on the lower energy side. Their intensities increased with the Sb beam flux applied during GaN growth. Since the central cell potential of tetrahedral-bonded Sb impurity is about 9 eV deep as compared to 14 eV of the N host atoms, the incorporated Sb atoms will act as isovalent traps in GaN epilayers. This is similar to the situation where Bi substitutes for P in GaP as an isoelectronic trap. 13 Isoelectronic trap has been used conventionally but was clearly a misnomer and therefore isovalent trap is used in our present article. In the theoretical description of isoelectronic traps or isovalent traps, the electrically neutral isoelectronic substitute must first trap one electronic particle, whereupon a second particle of opposite type can become bound in the Coulomb field of the first one to form the bound exciton BE state. The first particle is bound to the impurity atom by short-range non-Coulombic forces. A bound state can occur only if these forces are sufficiently strong, otherwise the isoelectronic impurity merely acts as a scattering center such as As substituting P in GaP.¹⁴ By considering the difference in electronegativities of the impurity atom and the atom it replaces in GaN, it is expected that Sb should be a neutral hole trap. When Sb atoms incorporate in GaN epilayers, holes and electrons will be trapped by these isovalent centers. Asgrown GaN without doping is always n type with a carrier concentration in the range of 10^{16} cm⁻³. ¹⁵ However GaN grown with Sb is highly resistive, indicating that most free carriers are trapped in the GaN layer. In photoluminescence spectra, the new transition at 3.27 eV was related to isovalent traps and its intensity increased with Sb beam flux applied in the GaN growth. It is conceivable that Sb incorporation in

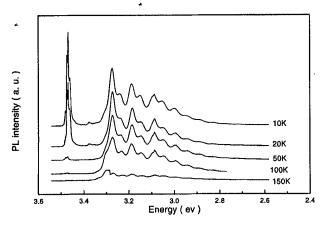


Fig. 4. Temperature-dependence photoluminescence spectra of GaN film grown with Sb.

GaN layers increased with the Sb beam flux applied in the GaN growth.

For a GaN sample grown with Sb beam equivalent pressure of 2.1×10^{-7} Torr, temperature-dependence PL was measured at different temperatures from 150 to 10 K. Excitonic emission was almost quenched at about 150 K, shown in Fig. 4. When the sample temperature was lowered, the intensities of both the excitonic emission and the transition related to Sb isovalent traps increased. Only at temperatures lower than 50 K could the phonon replica of the excitonic emission be observed.

IV. CONCLUSION

Growth of GaN on sapphire was dramatically improved with one monolayer Sb which was predeposited as surfactant. RHEED patterns became streaky much more rapidly when GaN was grown in the presence of Sb than that without Sb, indicating that Sb can act as an effective surfactant to

smooth the growth front of GaN and enhance the layer-bylayer growth mode for MBE growth of GaN. The FWHM of the 0004 XRD rocking curve of the GaN epilayer grown with one monolayer Sb as surfactant was as narrow as 70 arcsec. The influence of Sb on the photoluminescence of GaN was investigated. Transitions related to Sb isovalent traps were observed and their intensities increased with Sb beam flux.

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- ¹S. Nakamura, M. Senoh, and T. Mukai, Appl. Phys. Lett. **62**, 2390 1993.
- ²G. Feuillet, H. Hamaguchi, K. Ohta, P. Hacke, H. Okumura, and S. Yoshida, Appl. Phys. Lett. **70**, 1025 1997.
- ³Molecular Beam Epitaxy, Key Papers in Applied Physics, edited by A. Y. Cho AIP, New York, 1994.
- ⁴W. I. Wang, Appl. Phys. Lett. 44, 1149 1984.
- ⁵H. Kroemer, J. Cryst. Growth **81**, 193 1987.
- ⁶S. Nakamura, T. Mukai, and M. Senoh, J. Appl. Phys. 71, 5543 1992.
- ⁷Z. Yang, L. K. Li, and W. I. Wang, Appl. Phys. Lett. 67, 1686 1995.
- ⁸Z. Yang, L. K. Li, and W. I. Wang, J. Vac. Sci. Technol. B 14, 2354 1996.
- ⁹R. J. Molnar, R. Singh, and T. D. Moustakus, Appl. Phys. Lett. 66, 268 1995.
- ¹⁰X. Yang, M. J. Jurkovic, J. B. Héroux, and W. I. Wang, Electron. Lett. 35, 1082 1999.
- ¹¹X. Yang, J. B. Héroux, M. J. Jurkovic, and W. I. Wang, Appl. Phys. Lett. 76, 795 2000.
- ¹²F. Widmann, B. Daudin, G. Feuillet, N. Pelekanos, and J. L. Rouviere, Appl. Phys. Lett. 73, 2642 1998.
- ¹³J. J. Hopfield, D. G. Thomas, and R. T. Lynch, Phys. Rev. Lett. 17, 312 1966
- Japan, J. D. Cuthbert, and R. T. Lynch, Phys. Rev. 179, 754 1969.
 S. C. Jain, M. Willander, J. Narayan, and R. Van Overstraeten, J. Appl. Phys. 87, 965 2000.